WAK.094

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re patent application of

Toru Matsumoto

Serial No.: Not Yet Assigned — Group Art Unit: Not Yet Assigned

Filing Date: Concurrently Herewith — Examiner: Unknown

For: ELECTROCHEMICAL SENSOR

Assistant Commissioner of Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Prior to examination on the merits, please amend the above-identified application as follows:

IN THE SPECIFICATION:

Please replace the paragraph beginning at page 1, line 8 , with the following rewritten paragraph:

–When using a sensor performs an electrochemical measurement, a three-electrode method is widely employed, wherein an electrode group consisting of a working electrode, a counter electrode and a reference electrode is used. As an example of this type of sensor, a biosensor is disclosed in Figs. 1 (a)-1 (b) of Japanese Patent Application under Provisional Publication No. 256812/93, wherein a working electrode, a reference electrode and a counter electrode that are molded with platinum are provided on an aminoethanesulfonic insulating substrate. As another example, an electrochemical gas sensor is disclosed in Figs. 1(a)-1(b) and Fig. 6 of Japanese Patent Application under Provisional Publication No. 3323/94, wherein

two sets of a pair of electrode group consisting of a working electrode and a counter electrode are provided for a single reference electrode.--

Please replace the paragraph beginning at page 9, line 8, with the following rewritten paragraph:

--Figs. 1 (a)-1 (b) show a structure of a sensor relating to this invention.--

Please replace the paragraph beginning at page 9, line 10, with the following rewritten paragraph:

--Figs. 2 (a)-2 (b) show a structure of a sensor relating to this invention.--

Please replace the paragraph beginning at page 9, line 26, with the following rewritten paragraph:

--Figs. 9 (a)-9 (b) show a structure of a sensor relating to this invention.--

Please replace the paragraph beginning at page 10, line 8, with the following rewritten paragraph:

--Figs. 13 (a)-13 (c) show effects of interfering substances on an electric potential of a reference electrode.--

Please replace the paragraph beginning at page 10, line 10, with the following rewritten paragraph:

--Figs. 14 (a)-14 (b) show a structure of a sensor relating to this invention.--

Please replace the paragraph beginning at page 10, line 12, with the following rewritten paragraph:

--Figs. 15 (a)-15 (b) show the effects of an interfering substance on an output of a sensor and an electric potential of a reference electrode.--

Please replace the paragraph beginning at page 15, line 20, with the following rewritten paragraph:

--A structure of a sensor relating to the embodiment is shown in Figs. 1 (a)-1 (b) and a whole structure of a measuring apparatus including the sensor is shown in Fig. 3.--

Please replace the paragraph beginning at page 15, line 23, with the following rewritten paragraph:

--The sensor of the embodiment shown in Figs.1 (a)-1 (b) is a biosensor wherein a substance to be measured is converted to another substance by an enzyme reaction and the concentration of the converted substance is measured electrochemically. A working electrode 2, a counter electrode 3 and a reference electrode 4 is formed on an insulating substrate 1. An examining electrode 8 that measures and examines an electric potential of the reference

electrode 4 is formed on the substrate. On these electrode are formed a combining layer 20 (membrane thickness: about 10-50nm), an immobilized enzyme layer 21 (membrane thickness: about 200-1000nm) and a diffusion-limiting layer 22(membrane thickness: about 20-200nm) in this order. The wiring 5 may be an electric wire which can connects them. The cover 6 is placed so as to protect them.--

Please replace the paragraph beginning at page 23, line 11, with the following rewritten paragraph:

--The measuring apparatus consists of a sensor 10, a circuit of electrochemical measurement 11, a data processing unit 12 and a data indicator 13 and all of them are connected by a wiring 5. The sensor 10 has a structure as explained according to Figs. 1 (a)-1 (b). Since the sensor 10 is expendables, an exchangeable type by which replacement is easily performed is preferable. As the circuit of electrochemical measurement 11 a potentiostat is used in the embodiment of this invention, and there is no limitation to the circuit as far as the circuit can apply a constant electric potential to the sensor 10 and measure an electric current.--

Please replace the paragraph beginning at page 26, line 4, with the following rewritten paragraph:

--A structure of a sensor relating to the embodiment is shown in Figs. 2 (a)-2 (b). A basic structure of the sensor is nearly the same except for providing a spare reference electrode 9. The spare reference electrode 9 is used in place of the reference electrode 4 when an abnormal electric potential is detected in the reference electrode 4.--

Please replace the paragraph beginning at page 26 line 24, with the following rewritten paragraph:

--The examining electrode 8 is grounded and a voltmeter 17 is provided between the reference electrode 4 and the examining electrode 8 so that the examining electrode 8 can examine the electric potential of the reference electrode 4. The examining electrode 8 is connected to a means for switching the reference electrode which is not shown in Figs. 14 (a)-14 (b) and the means for switching the reference electrode performs the switching so that the spare electrode can be used when an abnormal electric potential is detected in the reference electrode 4.--

Please replace the paragraph beginning at page 27, line 26, with the following rewritten paragraph:

--A sensor shown in Figs. 1 (a)-1 (b) was made and the performance of the sensor was evaluated. A procedure of making the sensor is explained as follows:

A working electrode, a counter electrode, a reference electrode and an examining electrode were formed on a 10mm X 6mm quartz substrate. The working electrode (area: $7mm^2$) and the counter electrode (area: $4mm^2$) are made of platinum. The reference electrode and the examining electrode have a multi-layer structure of silver / silver chloride; at first silver membranes were formed on the substrate by the spattering method and then the substrate was dipped in an aqueous solution of iron chloride to form the reference electrode and the examining electrode.--

Please replace the paragraph beginning at page 30, line 9, with the following rewritten paragraph:

–In this example a sensor shown in Figs. 2 (a)-2 (b) was made and the performance of the sensor was evaluated. The procedure of making the sensor is explained as follows:

A working electrode, a counter electrode, a reference electrode and an examining electrode were formed on a 10mm X 6mm quartz substrate. The working electrode (area: $7mm^2$) and the counter electrode (area: $4mm^2$) are made of platinum. The reference electrode and the examining electrode have a multi-layer structure of silver / silver chloride; at first silver membranes were formed on the substrate by the spattering method and then the substrate was dipped in an aqueous solution of iron chloride to form the reference electrode and the examining electrode. - -

Please replace the paragraph beginning at page 32, line 19, with the following rewritten paragraph:

–In this example, a sensor shown in Figs. 9 (a)-9 (b) was made and a performance of the sensor was evaluated. A spare reference electrode 9 of this sensor is coated with a photoresist 15. The procedure of making the sensor is explained as follows:

A working electrode, a counter electrode, a reference electrode, an examining electrode and a spare reference electrode were formed on a 10mm X 6mm quartz substrate. The working electrode (area: $7mm^2$) and the counter electrode (area: $4mm^2$) are made of platinum. The reference electrode, the examining electrode and the spare reference electrode have a multi-layer structure of silver / silver chloride; at first silver membranes were formed by a spattering method and then the substrate was dipped in an aqueous solution of iron chloride to form the reference electrode, the examining electrode and the spare reference electrode.--

Please replace the paragraph beginning at page 36, line 12 with the following rewritten paragraph:

--As shown in Figs. 13 (a)-13 (c), an electrode potential in the sensor (ii) having no diffusion-limiting layer is greatly reduced by adding the interfering substance. On the other hand, it can be seen that a change of the electrode potential in the sensor (i) having the diffusion-limiting layer is controlled.

Reference Example 2.--

Please replace the paragraph beginning at page 37, line 1 with the following rewritten paragraph:

--The results of the measurement are shown in Figs. 15(a)-15(b). At first, the measured sample was a glucose solution that did not contain potassium sulfide and at the time indicated by an arrow 1mM of potassium sulfide was added. A sensor output and a reference electrode potential were changed by adding such a high concentration of an interfering substance. After the addition of the interfering substance the sensor still indicated the output but the value of the output continued to decrease as time elapsed.--

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REMARKS

The above changes to the specification have been made to correct the identity of the drawings.

Attached hereto is a marked-up version of the changes made to the specification by the current amendment. The attached pages are captioned **"<u>Version with markings to show changes made.</u>"**

Early, favorable prosecution on the merits is respectfully requested.

Please charge any deficiencies in fees and credit any overpayment of fees to Attorney's Deposit Account No. 50-0481.

Respectfully submitted,

46,060

for Sean M. McGinn
Registration No.: 34,386

Date: 8/8/01

McGinn & Gibb, PLLC
8321 Old Courthouse Road, Suite 200
Vienna, Virginia 22182-3817
(703) 761-4100
Customer No. 21254

VERSION WITH MARKINGS TO SHOW CHANGES MADE

In the specification:

Paragraph beginning at line 8 of page 1, has been amended as follows:

When using a sensor performs an electrochemical measurement, a three-electrode method is widely employed, wherein an electrode group consisting of a working electrode, a counter electrode and a reference electrode is used. As an example of this type of sensor, a biosensor is disclosed in [Fig.1] Figs. 1 (a)-1 (b) of Japanese Patent Application under Provisional Publication No. 256812/93, wherein a working electrode, a reference electrode and a counter electrode that are molded with platinum are provided on an aminoethanesulfonic insulating substrate. As another example, an electrochemical gas sensor is disclosed in [Fig.1] Figs. 1(a)-1(b) and Fig. 6 of Japanese Patent Application under Provisional Publication No. 3323/94, wherein two sets of a pair of electrode group consisting of a working electrode and a counter electrode are provided for a single reference electrode.

Paragraph beginning at line 8 of page 9, has been amended as follows:

[Fig.1 shows] Figs. 1 (a)-1 (b) show a structure of a sensor relating to this invention.

Paragraph beginning at line 10 of page 9 has been amended as follows:

[Fig.2 shows] Figs. 2 (a)-2 (b) show a structure of a sensor relating to this invention.

Paragraph beginning at line 26 of page 9 has been amended as follows:

[Fig.9 shows] Figs. 9 (a)-9 (b) show a structure of a sensor relating to this invention.

Paragraph beginning at line 8 of page 10 has been amended as follows:

[Fig.13 shows] Figs. 13 (a)-13 (c) show effects of interfering substances on an electric potential of a reference electrode.

Paragraph beginning at line 10 of page 10 has been amended as follows:

[Fig.14 shows] Figs. 14 (a)-14 (b) show a structure of a sensor relating to this invention.

Paragraph beginning at line 12 of page 10 has been amended as follows:

[Figs. 15 shows] Figs. 15 (a)-15(b) show the effects of an interfering substance on an output of a sensor and an electric potential of a reference electrode.

Paragraph beginning at line 20 of page 15 has been amended as follows:

A structure of a sensor relating to the embodiment is shown in [Fig.1] Figs. 1 (a)-1 (b) and a whole structure of a measuring apparatus including the sensor is shown in Fig. 3.

Paragraph beginning at line 23 of page 15 has been amended as follows:

The sensor of the embodiment shown in [Fig.1] Figs.1 (a)-1 (b) is a biosensor wherein a substance to be measured is converted to another substance by an enzyme reaction and the concentration of the converted substance is measured electrochemically. A working electrode 2, a counter electrode 3 and a reference electrode 4 is formed on an insulating substrate 1. An examining electrode 8 that measures and examines an electric potential of the reference

electrode 4 is formed on the substrate. On these electrode are formed a combining layer 20 (membrane thickness: about 10-50nm), an immobilized enzyme layer 21 (membrane thickness: about 200-1000nm) and a diffusion-limiting layer 22(membrane thickness: about 20-200nm) in this order. The wiring 5 may be an electric wire which can connects them. The cover 6 is placed so as to protect them.

Paragraph beginning at line 11 of page 23 has been amended as follows:

The measuring apparatus consists of a sensor 10, a circuit of electrochemical measurement 11, a data processing unit 12 and a data indicator 13 and all of them are connected by a wiring 5. The sensor 10 has a structure as explained according to [Fig.1] Figs. 1 (a)-1 (b). Since the sensor 10 is expendables, an exchangeable type by which replacement is easily performed is preferable. As the circuit of electrochemical measurement 11 a potentiostat is used in the embodiment of this invention, and there is no limitation to the circuit as far as the circuit can apply a constant electric potential to the sensor 10 and measure an electric current.

Paragraph beginning at line 4 of page 26 has been amended as follows:

A structure of a sensor relating to the embodiment is shown in [Fig.2] Figs. 2 (a)-2 (b). A basic structure of the sensor is nearly the same except for providing a spare reference electrode 9. The spare reference electrode 9 is used in place of the reference electrode 4 when an abnormal electric potential is detected in the reference electrode 4.

Paragraph beginning at line 24 of page 26 has been amended as follows:

The examining electrode 8 is grounded and a voltmeter 17 is provided between the reference electrode 4 and the examining electrode 8 so that the examining electrode 8 can examine the electric potential of the reference electrode 4. The examining electrode 8 is connected to a means for switching the reference electrode which is not shown in [Fig.14] Figs. 14 (a)-14 (b) and the means for switching the reference electrode performs the switching so that the spare electrode can be used when an abnormal electric potential is detected in the reference electrode 4.

Paragraph beginning at line 26 of page 27 has been amended as follows:

A sensor shown in [Fig.1] Figs. 1 (a)-1 (b) was made and the performance of the sensor was evaluated. A procedure of making the sensor is explained as follows:

A working electrode, a counter electrode, a reference electrode and an examining electrode were formed on a 10mm X 6mm quartz substrate. The working electrode (area: 7mm^2) and the counter electrode (area: 4mm^2) are made of platinum. The reference electrode and the examining electrode have a multi-layer structure of silver / silver chloride; at first silver membranes were formed on the substrate by the spattering method and then the substrate was dipped in an aqueous solution of iron chloride to form the reference electrode and the examining electrode.

Paragraph beginning at line 9 of page 30 has been amended as follows:

In this example a sensor shown in [Fig.2] Figs. 2 (a)-2 (b) was made and the performance of the sensor was evaluated. The procedure of making the sensor is explained as follows:

A working electrode, a counter electrode, a reference electrode and an examining electrode were formed on a 10mm X 6mm quartz substrate. The working electrode (area: 7mm^2) and the counter electrode (area: 4mm^2) are made of platinum. The reference electrode and the examining electrode have a multi-layer structure of silver / silver chloride; at first silver membranes were formed on the substrate by the spattering method and then the substrate was dipped in an aqueous solution of iron chloride to form the reference electrode and the examining electrode.

Paragraph beginning at line 19 of page 32 has been amended as follows:

In this example, a sensor shown in [Fig.9] Figs. 9 (a)-9 (b) was made and a performance of the sensor was evaluated. A spare reference electrode 9 of this sensor is coated with a photoresist 15. The procedure of making the sensor is explained as follows:

A working electrode, a counter electrode, a reference electrode, an examining electrode and a spare reference electrode were formed on a 10mm X 6mm quartz substrate. The working electrode (area: 7mm^2) and the counter electrode (area: 4mm^2) are made of platinum. The reference electrode, the examining electrode and the spare reference electrode have a multi-layer structure of silver / silver chloride; at first silver membranes were formed by a spattering method and then the substrate was dipped in an aqueous solution of iron chloride to form the reference electrode, the examining electrode and the spare reference electrode.

Paragraph beginning at line 12 of page 36 has been amended as follows:

As shown in [Fig.13] Figs. 13 (a)-13 (c), an electrode potential in the sensor (ii) having no diffusion-limiting layer is greatly reduced by adding the interfering substance. On the other

hand, it can be seen that a change of the electrode potential in the sensor (i) having the diffusion-limiting layer is controlled.

Reference Example 2.

Paragraph beginning at line 1 of page 37 has been amended as follows:

The results of the measurement are shown in [Fig.15] Figs. 15(a)-15(b). At first, the measured sample was a glucose solution that did not contain potassium sulfide and at the time indicated by an arrow 1mM of potassium sulfide was added. A sensor output and a reference electrode potential were changed by adding such a high concentration of an interfering substance. After the addition of the interfering substance the sensor still indicated the output but the value of the output continued to decrease as time elapsed.